

# Nonlocal explanation of stationary and nonstationary regimes in cascaded soliton pulse compression

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We study soliton pulse compression in materials with cascaded quadratic nonlinearities and show that the group-velocity mismatch creates two different temporally nonlocal regimes. They correspond to what is known as the stationary and nonstationary regimes. The theory accurately predicts the transition to the stationary regime, where highly efficient pulse compression is possible. © 2008 Optical Society of America

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Efficient soliton pulse compression is possible using second-harmonic generation (SHG) in the limit of large phase mismatch, because a Kerr-like nonlinear phase shift is induced on the fundamental wave (FW). Large negative phase shifts can be created, since the phase mismatch determines the sign and magnitude of the effective cubic nonlinearity. This induced *self-defocusing* nonlinearity thus creates a negative linear chirp through an effective self-phase modulation (SPM) term, and the pulse can therefore be compressed with normal dispersion. Beam filamentation and other problems normally encountered due to *self-focusing* in cubic media are therefore avoided. This self-defocusing soliton compressor can create high-energy few-cycle fs pulses in bulk materials with no power limit [1–4]. However, the group-velocity mismatch (GVM) between the FW and second harmonic (SH) limits the pulse quality and compression ratio. Especially very short input pulses ( $< 100$  fs) give asymmetric compressed pulses and pulse splitting occurs [4, 5]. In this case, the system is in the *nonstationary regime*, and conversely when GVM effects can be neglected it is in the *stationary regime* [3–5]. Until now, the stationary regime was argued to be when the characteristic GVM length is 4 times longer than the SHG coherence length [1], while a more accurate perturbative description showed that the FW has a GVM-induced Raman-like term [4, 5], which must be small for the system to be in the stationary regime [4]. However, no precise definition of the transition between the regimes exists.

On the other hand, the concept of nonlocality provides accurate predictions of quadratic spatial solitons [6, 7], and many other physical systems (see [8] for a review). Here we introduce the concept of nonlocality to the temporal regime and soliton pulse compression in quadratic nonlinear materials. As we shall show, GVM, the phase mismatch, and the SH group-velocity dispersion (GVD) all play a key role in defining the nonlocal behavior of the system. Two different nonlocal response functions appear naturally, one with a localized amplitude – rep-

resenting the stationary regime – and one with a purely oscillatory amplitude – representing the nonstationary regime. In the presence of GVM they are asymmetric and thus give rise to a Raman effect on the compressed pulse.

In the theoretical analysis we may neglect diffraction, higher-order dispersion, cubic Raman terms, and self-steepening to get the SHG propagation equations for the FW ( $\omega_1$ ) and SH ( $\omega_2 = 2\omega_1$ ) fields  $E_{1,2}(z, t)$  [3, 9]:

$$(i\partial_z - \frac{1}{2}k_1^{(2)}\partial_{tt})E_1 + \kappa_1 E_1^* E_2 e^{i\Delta kz} + \rho_1 E_1 (|E_1|^2 + \eta|E_2|^2) = 0, \quad (1)$$

$$(i\partial_z - id_{12}\partial_t - \frac{1}{2}k_2^{(2)}\partial_{tt})E_2 + \kappa_2 E_1^2 e^{-i\Delta kz} + \rho_2 E_2 (|E_2|^2 + \eta|E_1|^2) = 0, \quad (2)$$

where  $\eta = 2/3$  for Type I SHG [3].  $\kappa_j = \omega_1 d_{\text{eff}}/cn_j$ ,  $d_{\text{eff}}$  is the effective quadratic nonlinearity,  $\rho_j = \omega_j n_{\text{Kerr},j}/c$ , and  $n_{\text{Kerr},j} = 3\text{Re}(\chi^{(3)})/8n_j$  is the cubic (Kerr) nonlinear refractive index. The phase mismatch is  $\Delta k = k_2 - 2k_1$ , with  $k_j = n_j\omega_j/c$ .  $n_j$  is the refractive index, and  $k_j^{(n)} = \partial^n k_j / \partial \omega^n|_{\omega=\omega_j}$  accounts for dispersion. The time coordinate moves with the FW group velocity  $v_{g,1} = 1/k_1^{(1)}$ , giving the GVM term  $d_{12} = v_{g,1}^{-1} - v_{g,2}^{-1}$ . In dimensionless form,  $\tau = t/T_0$ , where  $T_0$  is the FW input pulse duration,  $\xi = z/L_{D,1}$ , where  $L_{D,1} = T_0^2/|k_1^{(2)}|$  is the FW dispersion length, and  $U_1 = E_1/\mathcal{E}_0$ ,  $U_2 = E_2/\sqrt{\bar{n}}\mathcal{E}_0$ , where  $\mathcal{E}_0 = E_1(0,0)$  and  $\bar{n} = n_1/n_2$ . Equations (1) and (2) become [3]

$$(i\partial_\xi - D_1\partial_{\tau\tau})U_1 + \sqrt{\Delta\beta}N_{\text{SHG}}U_1^*U_2e^{i\Delta\beta\xi} + N_{\text{Kerr}}^2U_1(|U_1|^2 + \eta\bar{n}|U_2|^2) = 0, \quad (3)$$

$$(i\partial_\xi - i\delta\partial_\tau - D_2\partial_{\tau\tau})U_2 + \sqrt{\Delta\beta}N_{\text{SHG}}U_1^2e^{-i\Delta\beta\xi} + 2\bar{n}^2N_{\text{Kerr}}^2U_2(|U_2|^2 + \eta\bar{n}^{-1}|U_1|^2) = 0, \quad (4)$$

with  $\delta = d_{12}T_0/|k_1^{(2)}|$ ,  $D_j = k_j^{(2)}/2|k_1^{(2)}|$ , and  $\Delta\beta = \Delta kL_{D,1}$ . The scaling conveniently gives the SHG soliton number [3, 4],  $N_{\text{SHG}} = L_{D,1}\mathcal{E}_0^2\omega_1^2d_{\text{eff}}^2/(c^2n_1n_2\Delta k)$ , and the Kerr soliton number  $N_{\text{Kerr}}^2 = L_{D,1}n_{\text{Kerr},1}\mathcal{E}_0^2\omega_1/c$ .

In the cascading limit  $\Delta\beta \gg 1$  the nonlocal approach takes  $U_2(\xi, \tau) = \phi_2(\tau) \exp(-i\Delta\beta\xi)$ , keeping its time dependence but neglecting the dependence on  $\xi$  of  $\phi_2$ . To do this the coherence length  $L_{\text{coh}} = \pi/|\Delta k|$  must be the shortest characteristic length scale in the system, which is true in all cascaded compression experiments. Discarding the Kerr terms in Eq. (4) because  $N_{\text{Kerr}}^2 U_2 \ll \sqrt{\Delta\beta} N_{\text{SHG}}$ , we get an ordinary differential equation  $D_2 \phi_2'' + i\delta \phi_2' - \Delta\beta \phi_2 = \sqrt{\Delta\beta} N_{\text{SHG}} U_1^2$ , with the formal solution

$$\phi_2(\tau) = -\frac{N_{\text{SHG}}}{\sqrt{\Delta\beta}} \int_{-\infty}^{\infty} d\tau' R_{\pm}(\tau') U_1^2(\tau - \tau'). \quad (5)$$

According to the sign of the parameter  $s_1 = \text{sgn}(\Delta\beta/D_2 - \delta^2/4D_2^2)$ ,  $R_+$  or  $R_-$  must be used

$$R_+(\tau) = \frac{\tau_2^2 + \tau_1^2}{2\tau_1\tau_2^2} e^{-is_2\tau/\tau_2} e^{-|\tau|/\tau_1}, \quad (6)$$

$$R_-(\tau) = \frac{\tau_2^2 - \tau_1^2}{2\tau_1\tau_2^2} e^{-is_2\tau/\tau_2} \sin(|\tau|/\tau_1), \quad (7)$$

where  $s_2 = \text{sgn}(D_2/\delta)$  and  $\int_{-\infty}^{\infty} d\tau R_+(\tau) = 1$ . Both response functions have an asymmetric imaginary part due to GVM, causing a Raman effect. Moreover,  $R_+$  is localized in amplitude (corresponding to the stationary regime), while  $R_-$  is purely oscillating in amplitude (corresponding to the nonstationary regime). The nonlocal dimensionless temporal scales are

$$\tau_1 = |\Delta\beta/D_2 - \tau_2^{-2}|^{-1/2}, \quad \tau_2 = 2|D_2/\delta|. \quad (8)$$

On dimensional form  $t_1 = \tau_1 T_0 = |2\Delta k/k_2^{(2)} - t_2^{-2}|^{-1/2}$ ,  $t_2 = \tau_2 T_0 = |k_2^{(2)}/d_{12}|$ , and  $\mathcal{R}_{\pm} = R_{\pm}/T_0$ , which all are independent of  $T_0$ . The transition between the stationary and nonstationary regimes occurs when  $s_1$  changes sign, which in dimensional units implies that

$$d_{12}^2 < 2\Delta k k_2^{(2)}, \quad \text{stationary regime, } R_+, \quad (9)$$

$$d_{12}^2 > 2\Delta k k_2^{(2)}, \quad \text{nonstationary regime, } R_-. \quad (10)$$

The transition is independent of  $T_0$  but depends on the GVM, the SH GVD, and the phase mismatch. This central result is important wherever cascaded quadratic nonlinear phase shifts are used. It should be compared with the qualitative arguments of [1], where the stationary regime was  $\Delta k < 4\pi|d_{12}|/T_0$ .

Now, inserting Eq. (5) into (3) gives a dimensionless generalized nonlinear Schrödinger equation

$$[i\partial_{\xi} - D_1\partial_{\tau\tau}]U_1 + N_{\text{Kerr}}^2 U_1|U_1|^2 - N_{\text{SHG}}^2 U_1^* \int_{-\infty}^{\infty} d\tau' R_{\pm}(\tau') U_1^2(\tau - \tau') = 0, \quad (11)$$

In the weakly nonlocal limit of the stationary regime  $\tau_{1,2} \ll 1$ ,  $U_1^2$  is slow compared with  $R_+$ , so

$$[i\partial_{\xi} - D_1\partial_{\tau\tau}]U_1 - (N_{\text{SHG}}^2 - N_{\text{Kerr}}^2)U_1|U_1|^2 = is_2 N_{\text{SHG}}^2 \tau_{R,\text{SHG}} |U_1|^2 \partial_{\tau} U_1, \quad (12)$$

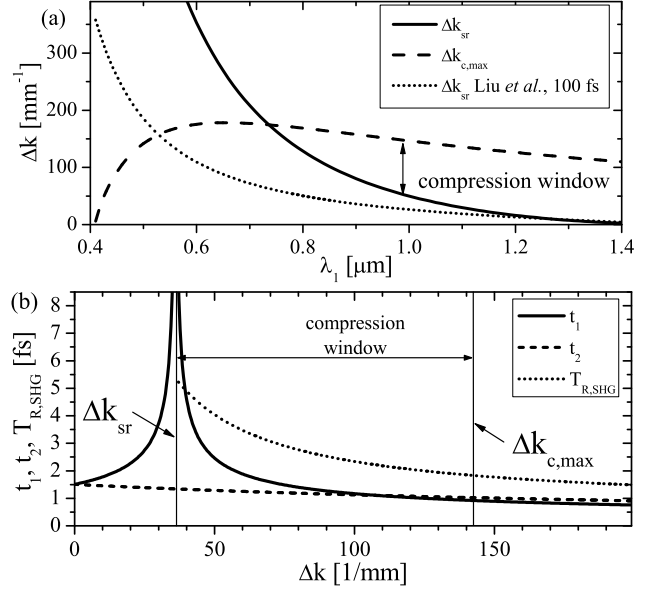


Fig. 1. (a) Compression window Eq. (14) versus  $\lambda_1$  in a BBO. Also  $\Delta k_{\text{sr}} = 4\pi|d_{12}|/T_0$  of [1] for a 100 fs pulse is shown. (b)  $t_{1,2}$  and  $T_{R,\text{SHG}}$  versus  $\Delta k$  for  $\lambda_1 = 1064$  nm.

where  $\int_{-\infty}^{\infty} dt R_+(t) = 1$  was used. The dimensionless characteristic time of the nonlocal Raman response is

$$\tau_{R,\text{SHG}} \equiv \frac{T_{R,\text{SHG}}}{T_0} = \frac{4\tau_1^2\tau_2}{\tau_1^2 + \tau_2^2} = \frac{2|d_{12}|}{\Delta k T_0} \quad (13)$$

Exactly this result has been derived before by perturbative methods [3–5]; that method therefore amounts to being in the weakly nonlocal limit of the stationary regime. Clearly, both Eq. (11) and (12) have terms reminiscent of the nonlinear Schrödinger equation with a purely cubic nonlinearity [10]; the GVM-induced nonlocality is similar to the Raman terms from a delayed cubic response.

From Eqs. (9) and (11) clean soliton compression requires, first, being in the stationary regime, i.e.,  $\Delta k > \Delta k_{\text{sr}} = d_{12}^2/2k_2^{(2)}$ . Second, soliton compression requires  $N_{\text{eff}} = \sqrt{N_{\text{SHG}}^2 - N_{\text{Kerr}}^2} > 1$  [3]. This can be expressed as  $\Delta k < \Delta k_c \equiv \Delta k_{c,\text{max}}/(1 + N_{\text{Kerr}}^{-2})$ , where  $\Delta k_{c,\text{max}} \equiv \omega_1 d_{\text{eff}}^2/n_{\text{Kerr},1}cn_1n_2$ . To remove the dependence on the FW input pulse intensity and duration it is convenient to require  $\Delta k < \Delta k_{c,\text{max}}$ , which is necessary to have  $N_{\text{eff}} > 1$ . Thus, we obtain a *compression window*:

$$\Delta k_{\text{sr}} < \Delta k < \Delta k_{c,\text{max}}. \quad (14)$$

In Fig. 1(a) we show the compression window for a  $\beta$ -barium-borate (BBO) crystal (see [3] for BBO material parameter details). Notice that the window closes for  $\lambda_1 < 750$  nm. Opening the compression window here requires a material with a stronger quadratic nonlinearity, or alternatively a strong dispersion control, as offered by photonic crystal fibers [11]. At  $\lambda_1 = 800$  nm the window

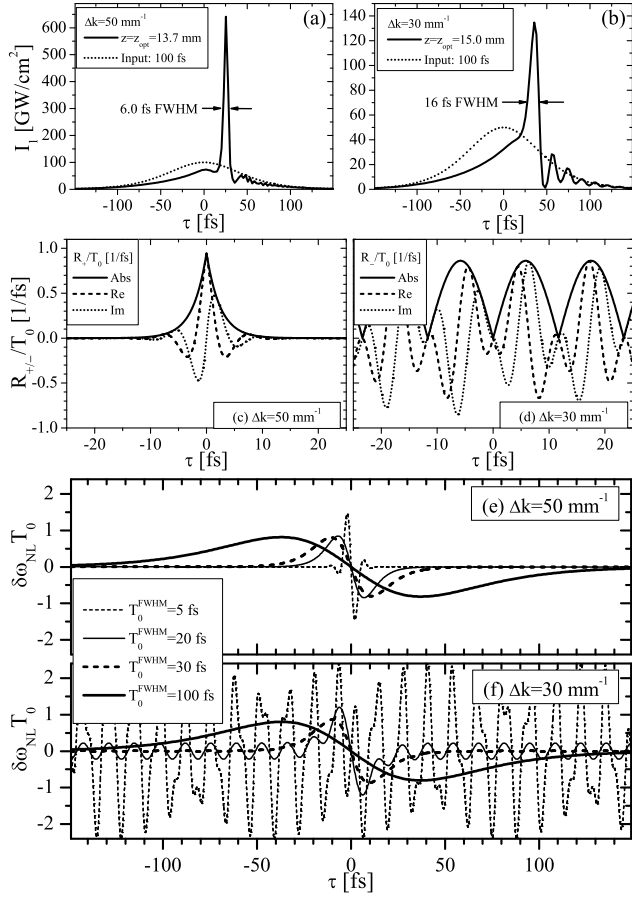


Fig. 2. Numerical simulations of soliton compression of a 100 fs sech pulse in BBO for  $\lambda_1 = 1064$  nm and  $N_{\text{eff}} = 5.3$ . The normalized FW intensity at  $z_{\text{opt}}$  is shown for (a)  $\Delta k = 50 \text{ mm}^{-1}$  (stationary regime) and (b)  $\Delta k = 30 \text{ mm}^{-1}$  (nonstationary regime). The response functions  $R_{\pm}/T_0$  and  $\delta\omega_{\text{NL}}T_0$  induced by cascaded SPM for (a) and (b) are shown in (c) and (e) and (d) and (f), respectively.

is narrow. In fact, the compression experiments done at 800 nm [1,2] were both in the nonstationary regime, and were both unable to observe compression to few-cycle pulses. Choosing  $\lambda_1 = 1064$  nm, Fig. 1(b) shows the nonlocal time scales versus  $\Delta k$ . At the transition to the nonstationary regime,  $t_1$  diverges while  $t_2$  remains low.

Let us illustrate the two regimes. Figures 2(a) and 2(b) show two numerical simulations of soliton compression of a 100 fs pulse at  $\lambda_1 = 1064$  nm in a BBO (the full coupled equations of [3] are solved). In Fig. 2(a)  $\Delta k = 50 \text{ mm}^{-1}$ ; our nonlocal theory predicts  $\Delta k_{\text{sr}} = 36.0 \text{ mm}^{-1}$ , so this is in the stationary regime. Indeed, a symmetric compressed 6 fs pulse is observed. Instead, changing to the nonstationary regime,  $\Delta k = 30 \text{ mm}^{-1}$  in Fig. 2(b), the pulse at  $z = z_{\text{opt}}$  (optimal compression point) is very asymmetric and strong pulse splitting occurs. Note, the definition in [1] predicts this simulation

to be in the stationary regime. The nonlocal response functions are shown in Figs. 2(c) and 2(d); while the nonlocal time scales are clearly quite similar for both examples, the different shapes of the response functions imply a very different impact on the pulse dynamics. This can be understood by using Eq. (11) to calculate the chirp  $\delta\omega_{\text{NL}}$  induced by SPM from the cascaded SHG process of a sech input pulse (calculations as in [10], Chap. 4). Figures 2(e) and 2(f) show  $\delta\omega_{\text{NL}}$  for  $z = L_{\text{SHG}}$  (where  $N_{\text{SHG}}^2 \equiv L_{\text{D},1}/L_{\text{SHG}}$  [3]). In the stationary case, Fig. 2(e), SPM induces a linear negative chirp on the central part of the pulse because  $R_+$  is localized, except for very short pulses,  $T_0 \sim t_1$ , where  $R_+$  becomes nonlocal. In the nonstationary case, Fig. 2(f), SPM induces a linear chirp only for long pulses  $> 30$  fs, while shorter pulses also have strong chirp induced in the wings because of the oscillatory character of  $R_-$ ; this explains the trailing pulse train in Fig. 2(b). Thus, the nonstationary response can be nonlocal even for  $T_0 \gg t_{1,2}$ . Note, in Fig. 2(b) very short pulses ( $T_0 \sim t_{1,2}$ ) are *positively* chirped in the central part (equivalent to a chirp induced by a self-focusing nonlinearity), making few-cycle compressed pulses impossible in the nonstationary case.

To conclude we showed that GVM induces asymmetric nonlocal Raman responses that accurately explain the stationary and nonstationary regimes in cascaded quadratic soliton compressors. The nature of the response functions and their degree of nonlocality relative to the input pulse length is vital for the resulting compression. The theory offers new insight into the physics of this soliton compressor.

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